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DETERMINATION OF OXYGEN IN SILICON AND SILICON CARBIDE BY ACTIVATION WITH 27.2 MeV ALPHA PARTICLES

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16. Abstract A destructive determination of O in Si and Si carbide by activation with accelerated alpha-particles by using the $^{16}O(alpha,d)^{18}F$ and $^{16}O(alpha,pn)^{18}F$ nuclear reactions (threshold 20.4 and 23.2 MeV) was developed. The Si samples, 30 mm in diameter and 1 mm thick, were polished on one side, and on the other (ground) side Ni was applied chemically and soldered with Pb to a water-cooled Cu substrate. Optical quartz standard was fixed from the other side. Si carbide samples were soldered to a substrate with In. The prepared samples were irradiated $\leq$ 2 hr in a cyclotron with a 27.2-MeV alpha-particle beam, beam current $\leq$ 10 μA; the standards were irradiated for 5 min at the beam current $\leq$ 0.1 μA. The upper layers, approximately 4 and 10 mg/cm^2 deep were removed from the Si and Si carbide samples, respectively, by grinding and the positron activity of $^{18}F(t_{1/2}$ 110 min) was measured by using a gamma,gamma-coincidence spectrometer with 2 NaI(Tl) crystals. For analysis of Si carbide, the activity decay curve of the samples was recorded (approximately 15 hr) to find the contribution of the positron activity of ^{64}Cu ($t_{1/2}$ 12.9 hr) which formed from Ni impurity on irradiation.

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DETERMINATION OF OXYGEN IN SILICON AND SILICON CARBIDE BY ACTIVATION WITH 27.2 MeV ALPHA-PARTICLES

A. P. Dolgolenko, N. D. Kornienko, and P. G. Litovchenko

A radioactive method using alpha-particles and He^3 ions is one of the most sensitive ways of determining oxygen content in silicon. /184*

The use of alpha-particles up to 40 MeV in energy in determining oxygen in silicon was first described in the study [1]; results of later investigations are in [2]. This study made use of a direct radioactivity method with chemical isolation of the activity of F^{18}. The sensitivity attained was $1.5 \cdot 10^{14}$ atoms/cm^3. He^3 ions with energy of approximately 10 MeV were used in determining oxygen in silicon and germanium in the study [3]. Oxygen in silicon was determined in this investigation without chemical isolation; the sensitivity attained was $3 \cdot 10^{13}$ atoms/cm^3. The last method cannot serve in determining oxygen in carbon-containing materials without radiochemical isolation of fluorine. In this case, the interfering reaction $C^{12}(He^3, alpha)C^{11}$ has a low threshold (approximately 3.5 MeV); the reaction cross-section is as high as 320 mbarns when the He^3 ion energy is 10 MeV [4]. alpha-Particles with 40 MeV energy are also poorly suited for this purpose, since the activation threshold of carbon is 24.9 MeV and the cross-section is about 70 mbarns at 40 MeV [2].

We developed a method of determining oxygen in silicon and in silicon carbide by their activation with 27.2 MeV alpha-particles, without radiochemical isolation. The irradiation was conducted on a cyclotron at the Institute of Physics, Ukrainian

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* Numbers in the margin indicate pagination in the foreign text.

SSR Academy of Sciences. The oxygen content was determined from the activity of F^{18} formed in the reactions

$O^{16}(\alpha, d)F^{18}$; $O^{16}(\alpha, pn)F^{18}$;

$$E_{th} = 20.4 \text{ MeV} \quad (1)$$

$$E_{th} = 23.2 \text{ MeV} \quad (2)$$

In silicon carbide, because of differences in the thresholds of reactions (1) and (3) with respect to oxygen and carbon

$$C^{12}(\alpha, \alpha n)C^{11}, \quad E_{th} = 24.9 \text{ MeV} \quad (3)$$

oxygen can be determined without radiochemical isolation of F^{18}. /185

The positron activity of F^{18}, which has a half-life of 110 min, was recorded with a gamma-gamma-coincidence spectrometer, with an efficiency of about 4 percent, against a background of 1.7 coincidences per minute. The spectrometer consists of two photomultipliers equipped with 80x80 mm NaI(Tl) crystals with a 17 percent resolution with respect to Cu^{64}, two identical analyzers, and a coincidence circuit with a resolving time of 0.6 μsec.

Before irradiation, the silicon specimens, up to 30 mm in diameter and 1 mm in thickness, were polished on one side; nickel was chemically deposited on the other (polished) side. For good heat removal, the specimens were soldered with tin to a water-cooled copper substrate. The silicon carbide specimens were soldered to the substrate with indium. Standards were attached to the other side. Optical quartz (ρ = 2.2 g/cm^3) was the standard. The specimens were irradiated at a current to 10 μA; the irradiation time did not exceed 2 hr; the standards were irradiated for 5 min at current values to 0.1 μA.

In calculating the oxygen content in silicon and silicon carbide specimens, we used a formula derived under the following assumptions: 1) the range of the alpha-particles, expressed in

milligrams per square centimeter, in silicon and silicon carbide is equal to the range of alpha-particles in quartz; 2) the reduction in the alpha-particle flux owing to nuclear reactions was not taken into account.

The formula is as follows:

$$n = \left[N \frac{R_{SiO_2}}{R_x}\right] \frac{A_x i_{SiO_2} (1 - e^{-\lambda t_{SiO_2}}) e^{-\lambda t'_{SiO_2}}}{A_{SiO_2} i_x (1 - e^{-\lambda t_x}) e^{-\lambda t'_x}},$$

where n is the concentration of oxygen atoms per cm^2 of test specimen; R_{SiO_2}, R_x are the ranges of alpha-particles in the standard (silicon oxide) and in the test specimen, cm; N is the concentration of the oxygen atoms per cm^3 of the standard; A_x and A_{SiO_2} is the activity of F^{18} in the specimen and in the standard at the time of measurement; i_x, i_{SiO_2} is the He^4 ion current recorded from the test specimen and from the standard; t_{SiO_2}, t_x are the irradiation times of the standard and the test specimen; t'_{SiO_2}, t'_x are the times from the beginning of the end of irradiation to the beginning of measurement for the standard and for the specimen; λ is the decay constant of the F^{18} nucleus.

The expression in the brackets, for silicon carbide, let us say, can be written as follows:

$$N \frac{R_{SiO_2}}{R_x} = 2N_{SiC} \frac{Z_{Si} + Z_C}{Z_{Si} + 2Z_O} = N \frac{\rho_{SiC}}{\rho_{SiO_2}},$$

where N_{SiC} is the molecular concentration of the test specimen (SiC) per cm^3; Z_{Si} is the atomic number of silicon; Z_O is the atomic number of oxygen; Z_C is the atomic number of carbon; /186 ρ_{SiC}, ρ_{SiO_2} are the specific densities of silicon carbide and silicon oxide. When we calculated the oxygen content in silicon, we also employed Eq. (1) from the study [3].

Oxygen content was determined in the polished layers of the silicon monosilane specimens (Figures 1 and 2) by comparing the

activity of the polished layers of the standard and of the specimen. The oxygen content in these specimens ranges from $1.6 \cdot 10^{15}$ to $5.5 \cdot 10^{15}$ atoms/cm^3. The measured oxygen concentration in the silicon specimen grown by the Czochralski method is $1.5 \cdot 10^{18}$ atoms/cm^3; this is nearly twice as high as the data from the infrared absorption method.

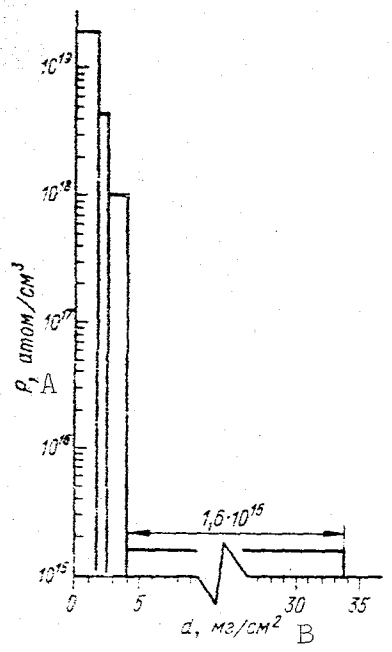

Figure 1. Distribution of oxygen content in silicon ($\rho = 4 \cdot 10^3$ ohm·cm) over the depth of the irradiated specimen

Key: A. atoms/cm^3
B. mg/cm^2

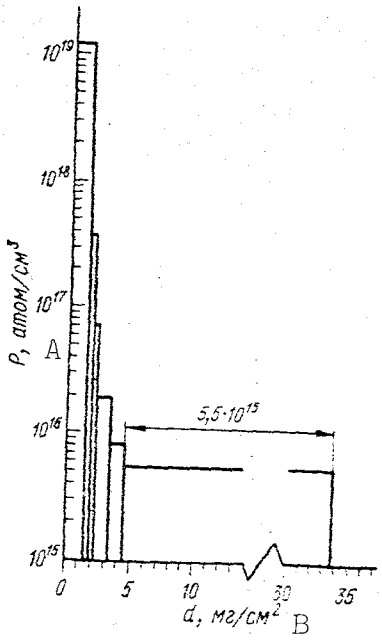

Figure 2. Distribution of oxygen content in silicon ($\rho = 2.6 \cdot 10^4$ ohm·cm) over the depth of the irradiated specimen

Key: A. atoms/cm^3
B. mg/cm^2

The resulting distributions of oxygen content in the irradiated silicon show that the depth of the removed layer must be

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approximately 4 mg/cm^2 for an accurate determination of oxygen content in silicon specimens.

To avoid the positron activity of C^{11} in the reaction C^{12}(alpha,alpha-n)C^{11} with silicon carbide specimens, a layer approximately 10 mg/cm^2 was polished off with diamond paste, then the decay curves of specimen activity were recorded. A /187
typical decay curve of silicon carbide specimen activity, after the removal of an 8 mg/cm^2 layer, is shown in Figure 3. In the figure we see that the decay curve consists of two components with half-decay times of 110 min and 12.9 hr. The first component corresponds to the decay of F^{18}, and the second--the decay of C^{64}, formed most probably from Ni^{61} and Ni^{62} (nickel impurities in silicon carbide). The oxygen content in the silicon carbide specimens that were green in color ranged from $4.5 \cdot 10^{18}$ to $2 \cdot 10^{19}$ atoms/cm^2. In experiments with the irradiation of the standard (SiO_2) with 27.2 MeV alpha-particles and with the measurement of F^{18} activity, the sensitivity of oxygen determination in silicon and in silicon carbide was found. The sensitivity of this method was established under the following assumptions:

1) for reliable identification of F^{18}, the measuring time must be of the order of the half-life (2 hr)

2) the time for measuring a point on the decay curve is 30 min

3) the contribution from the interfering reaction F^{19}(alpha, alpha-n)F^{18} need not be considered, since under the technology of preparing silicon monosilane this impurity has a low probability of appearing; the activity of C^{11} after a 4 mg/cm^2 silicon layer was removed was not observed

4) by the end of measurements, the necessary excess of the count over the background was taken to be 3σ (σ is the standard deviation of the background). The background of our unit was 1.7 pulses/min

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5) the time elapsing between the end of irradiation and the beginning of measurements was 30 min for silicon and 3 hr for silicon carbide (the latter value was used in order to lessen the effect of C^{11} activity prior to polishing, although this time can be reduced to 30 min).

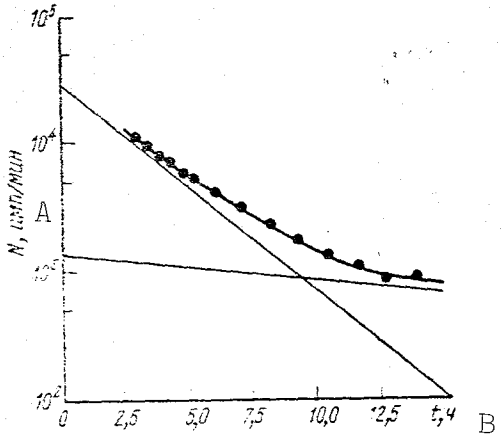

Figure 3. Decay curve of silicon carbide activity after polishing of an 8 mg/cm^2 layer. Light lines correspond to the decay curves of F^{18} and Cu^{64} activity.
Key: A. pulses/min B. hr

From the data for the irradiation of the standards, it was found that an oxygen content of approximately $1.5 \cdot 10^{14}$ atoms/cm^3 corresponds to 1 coincidence per minute.

Thus the sensitivity of oxygen determination in silicon was $3 \cdot 10^{14}$ atoms/cm^3 --under the above assumptions--after 2 hr irradiation and with a 10 μA current in the specimen, following removal of a 4 mg/cm^2 /188 surface silicon layer. The reproducibility of the results is approximately 30 percent.

Let us find the sensitivity of oxygen determination in silicon carbide. The ratio of the alpha-particle ranges in silicon and in silicon carbide can be written as follows:

$$\frac{R_{SiC}}{R_{Si}} = \frac{\rho_{Si} Z_{Si} A_{SiC}}{\rho_{SiC} Z_{SiC} A_{Si}} = 0{,}81,$$

where R_{SiC}, R_{Si} are the ranges in SiC and Si, cm; A_{SiC}, A_{Si} are the molecular and atomic weights of SiC and Si, respectively.

When alpha-particles pass through a 10.5 mg/cm^2 silicon layer, their energy falls off to the carbon activation threshold (24.9 MeV). The mean activation cross-section for oxygen in silicon after a 4 mg/cm^2 layer was removed was found to be approximately 16 mbarns; after a 10.5 mg/cm^2 layer was removed, it was determined to be about 10 mbarns. Therefore, the sensitivity of oxygen determination in silicon carbide is $8 \cdot 10^{14}$ $atoms/cm^3$.

Thus, the method developed lets us determine oxygen in silicon carbide at fairly high sensitivity. This method can serve in the determination of oxygen without chemical isolation of F^{18} activity in all carbide materials that are crystalline.

REFERENCES

1. Nozaki, T. et al., Nature, 190, 39 (1961).

2. Saito, K. et al., Int. J. Appl. Rad. Isotopes, 14/7, 357 (1963).

3. Aleksandrova, G. I. et al., Atomnaya energiya, 23, 106 (1967).

4. Markowitz, S. S., and Mahony, J. D., Radiochemical Methods of Analysis, Vol. 1, Vienna, IAEA, 1965, p. 419.